

Fourteen years of palaeolimnological research of a past industrial polluted lake (L. Orta, Northern Italy): an overview

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ABSTRACT

The first sediment core analyses were carried out in 1958, but it is only from 1985 onward that a modern palaeolimnological approach was applied to the study of Lake Orta, a large and deep lake in North Italy, heavily polluted by ammonia and copper for about 50 and 30 years, respectively. Thus, we summarise those studies from a variety of sediment cores, collected in different years and sites, using both published and unpublished data. Changes in algal pigments (mainly chlorophyll derivatives, total and single carotenoids), inorganic geochemistry, especially heavy metals (e.g. Cu), diatoms, Cladocera, and thecamoebians were studied and related to the stressed environment. The whole picture reveals a close relationship between modifications in algal biomass, density, taxonomic composition and organisms size on the one hand, and water chemistry changes on the other hand. Long-term history of this lake over 7-8 centuries, including invertebrate and terrestrial vegetation dynamics (pollen stratigraphy), reveals close relationship with natural (climate) and anthropogenic forces.

Key words: palaeolimnology, sediments, fossil remains, pigments, geochemistry, pollen, pollution

1. INTRODUCTION

Lake Orta is one of the most intensively studied freshwater environments in the world and the present collection of papers is only the last of a long series of publications (see, for example, papers quoted in Bonacina & Bonomi 1984; Bonacina *et al.* 1990; Calderoni & de Bernardi 1992). Long-term limnological data dating back to the beginning of 1900 has shown in great detail the dramatic history of its pollution by heavy metals and ammonium sulphate, events that have made this large, deep and important Italian water resource internationally renowned among limnologists.

Historical data concerning Lake Orta have also been obtained using palaeolimnological techniques. This approach was essential for determining the record of the pre-industrial centuries, but has also been invaluable for some recent periods (e.g., 1930-1950) in which field data were lacking or (e.g., phytoplankton) very scarce and incomplete. These gaps were first investigated by Corbella *et al.* (1958) in their pioneering work on sediment cores from Orta in which the historical record of the lake was reconstructed from geochemical records (e.g., Cu, organic content), sub-fossil diatom remains and plant pigments in the sediments. The results of this palaeolimnological study were in good agreement with the limnological data and showed clearly the strong impact of the heavy pollution by copper and ammonia on the lake chemistry and biota.

Further studies have subsequently been carried out on a number of sediment cores collected since 1978

(Tab. 1). The main objectives were: (1) to determine a detailed history of primary production (Adams *et al.* 1978; Guilizzoni *et al.* 1992, 1993); (2) to reconstruct changes in algal assemblages using species or group specific carotenoids (Guilizzoni & Lami 1988, 1990), and (3) to estimate the natural variability and impact of pollution on diatom, Cladocera (Ruggiu *et al.* 1998; Manca & Comoli 1995) and Oligochaete communities (Bonacina *et al.* 1986). Finally, a recently a study was published on the effects of pollutants on body size of three communities from different kingdoms and trophic levels (diatoms, Cladocera and thecamoebians; Cattaneo *et al.* 1998). Studies of mineralogy, lithology, pollen, magnetic properties (Alvisi 1993; Frignani *et al.* 1995; Alvisi *et al.* 1996) and sediment contamination by heavy metals (Provini & Gaggino 1985; Baudo *et al.* 1989; Guilizzoni & Lami 1990; Rossi *et al.* 1998), were also carried out.

The objective of the present paper is to present a general review of the main findings and results of these earlier studies (see Tabs 1-3 for a summary), and also to report new data on sedimentation rates, geochemistry, mineralogy, pollen and pigments for a more recent core collected in 1992.

2. SITE HISTORY

Lake Orta is a large deep subalpine lake in Northern Italy situated between 45°46' – 45°52' N and 8°23' – 8°26' W. It has a surface area of 18.5 km², a maximum depth of 143 m, a mean depth of 71 m and a volume of 1.3 km³ (Fig. 1). The catchment area consists primarily

Tab. 1. Core inventory for Lake Orta since the first study in 1956.

Core	Site	Date	Coring depth	Corer	Reference
OR-56/1	Buccione	1956	20 m	Moore	Corbella <i>et al.</i> 1958
OR-56/2	Buccione	1956	35 m	Moore	Corbella <i>et al.</i> 1958
OR-82/3	Buccione	1982	n.a.	Jenkin	Provini & Gaggino 1985
OR-85	Buccione	1985	30 m	Jenkin	Ruggiu <i>et al.</i> 1998
OR1	Buccione	1990	30 m	Mackereth	Alvisi 1993 ;Frignani <i>et al.</i> 1995
OR-94	Buccione	1994	30 m	Limnos	Cattaneo <i>et al.</i> 1998; Manca & Comoli 1995
OR-95	Buccione	1995	10 m	Scuba diving	Rossi <i>et al.</i> 1998
OR-56/4	Pettenasco	1956	122 m	Moore	Corbella <i>et al.</i> 1958
OR-78	Pettenasco	1978	100 m	Freeze corer	Adams <i>et al.</i> 1978
OR-85/B	Pettenasco	1985	~100 m	Jenkin	Bonacina <i>et al.</i> 1986
OR-86	Pettenasco	1986	120 m	Jenkin	Guilizzoni & Lami 1988
OR-92	Pettenasco	1992	120 m	gravity	this paper

Tab. 2. Major changes in sediment profiles (cm): (*) ²¹⁰Pb dating (this paper), (**) H. Muntau (unpublished data).

Event Date	Floods 1963, 1965	pigments peak	Cu peak 1958-'60	biota change 1928-'30	lithology change 1928-'30	LOI increase 1928-'30	Cu onset 1928-'30
Core							
OR-56/1				4			4
OR-56/2				4			4
OR-78		8-10			10	10	
OR-82/3			6-7				9
OR-85	4-5			9		10-11	
OR1	5 cm (*)		5.5 (*)		10	10	8 (*)
OR-94				7		7	
OR-95			6-8				
OR-56/4				5			6
OR-85/B							
OR-86	6-7	9-10	11			12	13
OR-92	5-8	10-11	11 (**)	13	12.5	12.5	12.5

Tab. 3. Mean sedimentation rates (cm y⁻¹) during the last 60 years at two coring sites, based on an historical flood event and on chemical and biological markers (cf. Tabs 1 & 2).

Core	since 1964	based on	since 1959	based on	since 1929	based on
at Buccione						
OR-56/1					0.15	Cu & biota
OR-56/2					0.15	Cu & biota
OR-82/3			0.28	Cu	0.17	Cu & biota
			0.24	¹³⁷ Cs		
OR-85	0.11	flood			0.19	LOI
					0.16	biota
OR1	0.14	²¹⁰ Pb	0.21	²¹⁰ Pb	0.13	²¹⁰ Pb
					0.16	biota & LOI
OR-94					0.11	biota & LOI
OR-95			0.19	Cu	0.09	Cu
at Pettenasco						
OR-56/3					0.22	biota
					0.19	LOI
OR-78			≤0.20	¹³⁷ Cs	0.20	LOI & pigments
OR-85/B					0.21	biota
OR-86	0.22	flood	0.41	Cu	0.21	LOI
					0.23	Cu
OR-92	0.15	flood	0.33	Cu	0.20	Cu & LOI

of acid rocks (granite, gneiss) and especially on the western side is extensively covered by woods of *Castanea sativa*, *Quercus* spp., *Juglans regia* and *Fagus sylvatica*. Further details are given in Frignani *et al.* (1995) (site description), Calderoni *et al.* (1999) (hydrochem-

istry), Alvisi (1993) (regional physiography and geology) and other papers in this issue.

Water quality deteriorated significantly after 1926 when a rayon factory (Fig. 1) began to discharge large amounts of copper (80 t y⁻¹) and ammonium sulphate

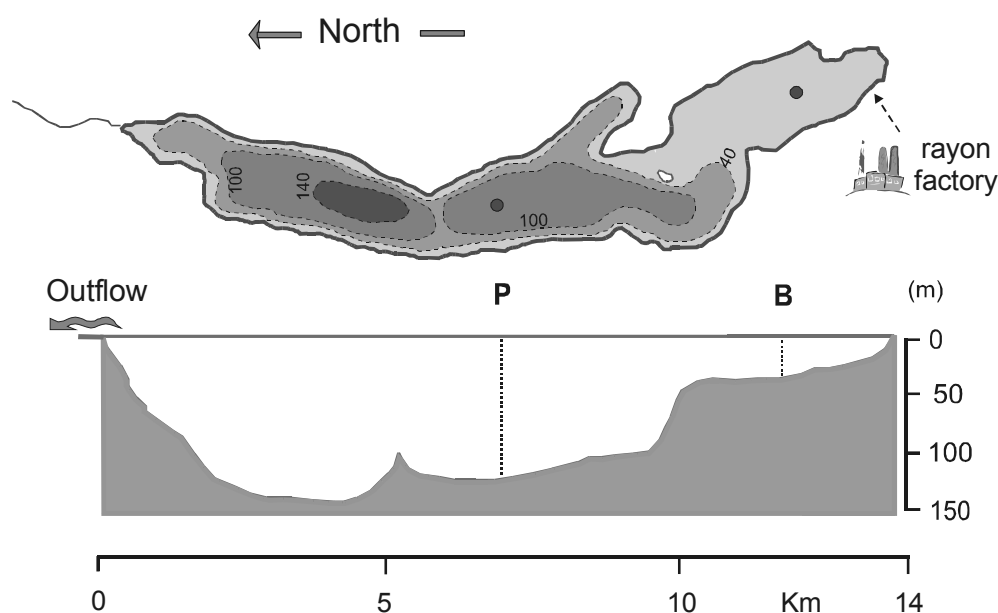


Fig. 1. Morphometry and longitudinal profile of Lake Orta: **P** (Pettenasco) and **B** (Buccione) are the coring sites. The location of the rayon factory responsible of the lake pollution for *ca* 50 years (1927-1980) is also indicated.

Tab. 4. ^{210}Pb chronology for core OR1 (Buccione) of Lake Orta.

Depth cm	Dry Mass (g cm^{-2})	Date AD	Age (y)	Sedim. Rate ($\text{g cm}^{-2} \text{y}^{-1}$)	Sedim. Rate (cm y^{-1})	\pm (%)
0.00	0.00	1990	0	0.038	0.27	4.9
1.00	0.09	1988	2 ± 2	0.034	0.23	5.2
2.00	0.25	1982	8 ± 2	0.024	0.19	5.5
3.00	0.40	1976	14 ± 2	0.019	0.16	5.8
4.00	0.56	1971	19 ± 2	0.015	0.12	6.2
5.00	0.72	1965	25 ± 2	0.015	0.093	7.8
6.00	0.89	1952	38 ± 2	0.015	0.093	7.8
7.00	1.06	1941	49 ± 3	0.015	0.093	7.8
8.00	1.23	1930	60 ± 3	0.015	0.093	7.8
9.00	1.39	1919	71 ± 4	0.015	0.093	7.8
10.00	1.55	1908	82 ± 5	0.015	0.093	7.8
11.00	1.71	1891	93 ± 6	0.015	0.087	-10
12.00	1.90	1885	105 ± 8	0.015	0.087	-10
13.00	2.08	1873	117 ± 10	0.015	0.087	-10
14.00	2.26	1861	129 ± 12	0.015	0.087	-10
15.00	2.44	1849	141 ± 14	0.015	0.087	-10
16.00	2.62	1837	153 ± 16	0.015	0.087	-10

(max $3350 \text{ t N-NH}_4 \text{ y}^{-1}$; Calderoni & Mosello 1990) into the lake. By 1950 the naturally poorly buffered waters in the profundal layers had become progressively acid (pH *ca* 4) and anoxic due to NH_4 in-lake oxidation. Concentrations of toxic substances in effluents from the rayon factory were greatly reduced in 1958 (Cu) and 1980 (NH_4), though a further source of pollution during the past 20-30 years has been from trace metals (e.g., Cr, Zn, Ni, Zn) in effluents from the many electroplating factories bordering the lake. In 1988 the lake was treated with lime (Calderoni *et al.* 1991) and since then has been progressively recovering its "stability" (Calderoni & Tartari 2001).

3. CHRONOLOGY AND SEDIMENT ACCUMULATION

A sediment core from Lake Orta (OR1; cf. Tab. 1), collected in July 1990 from the Buccione Basin (Fig. 1), was measured non-destructively in the Liverpool University Environmental Radiometric Laboratory for ^{210}Pb , ^{226}Ra , ^{241}Am , ^{137}Cs and ^{134}Cs by direct gamma assay using Ortec HPGE GWL series (Appleby *et al.* 1986; Appleby *et al.* 1992). The results of the radiometric analyses are given in table 4 and shown graphically in figures 2-4. Activity of the short-lived radiocaesium isotope ^{134}Cs , which derives solely from fallout from the

Chernobyl reactor accident, has been corrected for decay since 5th May 1986.

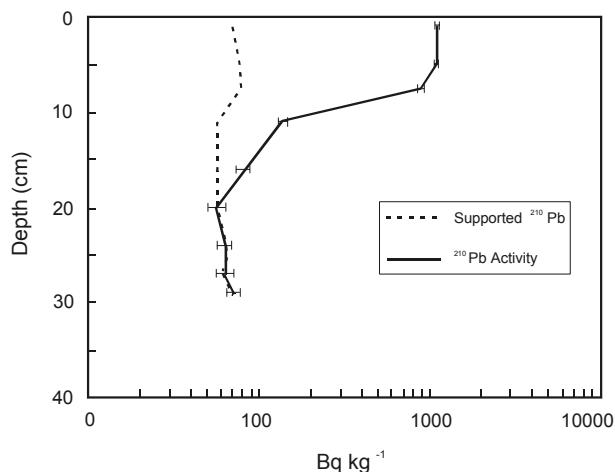


Fig. 2. Total and supported ^{210}Pb stratigraphy in Lake Orta core OR1.

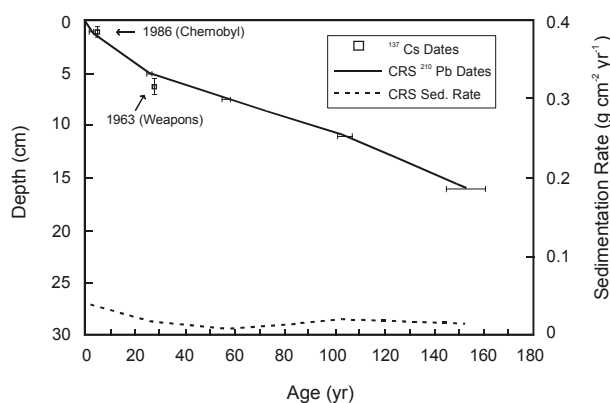


Fig. 3. Age-depth relationship and accumulation rate in Lake Orta core OR1 as determined by ^{210}Pb (Constant Rate of Supply model).

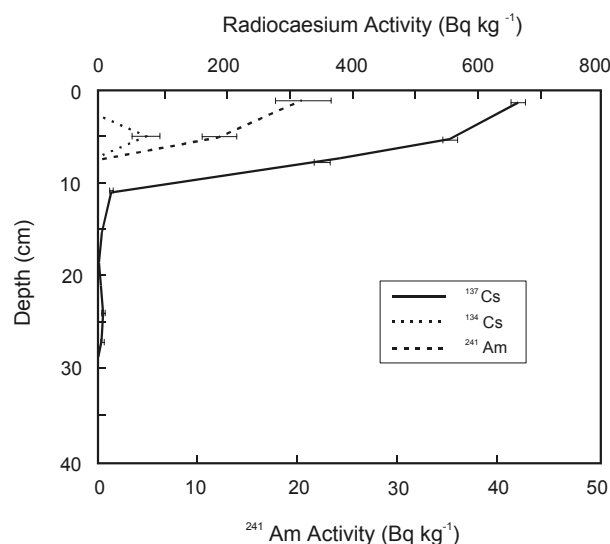


Fig. 4. Depth profiles of radiocaesium and ^{241}Am concentrations in Lake Orta core OR1.

Equilibrium between total ^{210}Pb activity and the supporting ^{226}Ra , corresponding to about 150 years of sediment accumulation, was reached at a depth of between 16–20 cm. Unsupported ^{210}Pb activity was virtually constant in the top 6 cm of the core but beneath this zone fell steeply following a more or less exponential relationship with depth.

The high ^{137}Cs and ^{134}Cs concentrations in the uppermost samples record deposition from the Chernobyl accident, and also confirm the integrity of the surficial sediment record. From the distribution of the Chernobyl fallout it is estimated that sediments from 1986 lie at a depth of not more than 2 cm. It is evident from the ^{134}Cs record that Chernobyl ^{137}Cs has diffused down the sediment column to a depth of *ca* 5–7 cm. Using the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio characteristic of Chernobyl fallout it was possible to partition the total ^{137}Cs activity into its Chernobyl and weapons fallout components and so reconstruct the weapons fallout ^{137}Cs record. The results of these calculations had large uncertainties, but nonetheless place the 1963 weapons fallout peak at a depth of between 5–7 cm. This was supported by the presence of traces of ^{241}Am at 5 cm depth (Appleby *et al.* 1991). The weapons ^{137}Cs record has a small but significant secondary peak in activity at 23–28 cm. Since this is well below the possible 1954 depth marking the onset of ^{137}Cs fallout the most likely cause is downwards transport of ^{137}Cs by pore water diffusion with preferential adsorption due to a change in mineralogy or sediment chemistry at these depths.

^{210}Pb dates calculated using the CRS (Constant Rate of Supply) model (Appleby & Oldfield 1978), shown in figure 3, are in relatively good agreement with the radiocaesium dates, implying that the near uniform ^{210}Pb activity in the surficial sediments is due to dilution of the constant atmospheric ^{210}Pb flux by accelerating sedimentation. The alternative explanation of physical mixing is precluded by the decline in radiocaesium concentrations in the top 6 cm of the core. Further, use of a mixing model would give a date for the weapons fallout peak that is clearly too old (1927 AD).

The ^{210}Pb dates for OR1, given in table 4, put 1930 at a depth of 8 cm, 1900 at a depth of 10.5 cm, and 1850 at a depth of 15 cm. Prior to 1971 there appears to have been a more or less constant sedimentation rates of $0.015 \pm 0.002 \text{ g cm}^{-2} \text{ y}^{-1}$ (*ca* 0.087 cm y^{-1}). Changes since then have more than doubled this to a present day value of about $0.038 \text{ g cm}^{-2} \text{ y}^{-1}$. The mean post-1963 sedimentation rate calculated from the ^{137}Cs dates is $0.033 \pm 0.006 \text{ g cm}^{-2} \text{ y}^{-1}$ (0.22 cm y^{-1}) compared to $0.029 \pm 0.003 \text{ g cm}^{-2} \text{ y}^{-1}$ (0.20 cm y^{-1}) by ^{210}Pb . These results compare quite well (especially for the last 40 years) with independent dating evidence from a number of cores from the Buccione basin using LOI data, pigments, biota assemblage changes and flood events (Tab. 3). Table 3 also shows that sedimentation rates in the Buccione Basin are lower than those in the deeper Pet-

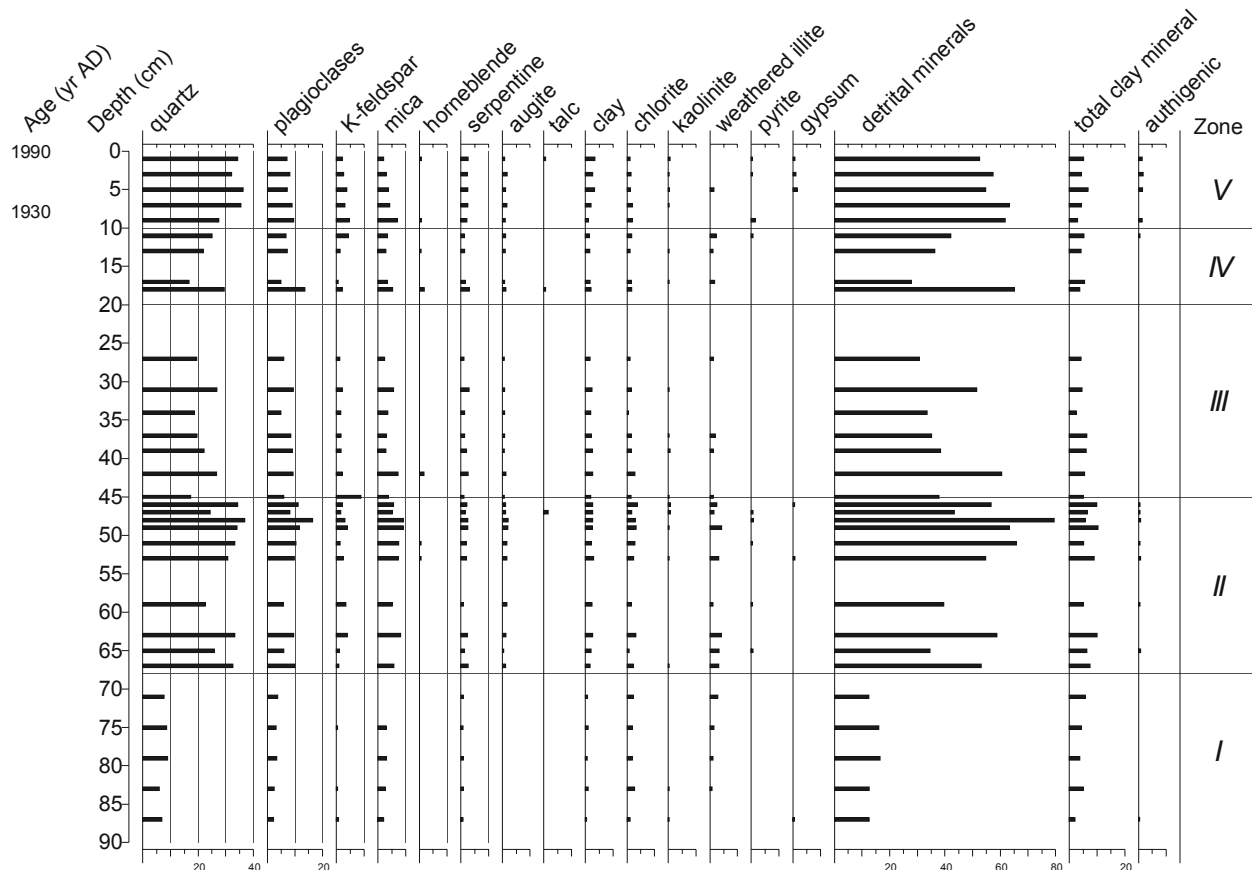


Fig. 5. Mineralogical composition (as absolute values of the heights of the measured peaks) of Lake Orta core OR1. Zone I-V are also indicated.

tenasco basin (e.g., core OR-92, results for which based on pigment analysis, LOI and other parameters are presented below for the first time).

4. CORE DESCRIPTION

The lithology of core OR1 can be subdivided in two main units. The deeper sediments (from the base of the core up to about 15 cm) were composed of a rather homogeneous brown to light-grey clay (ca 60%), silt (37%) and sand (<1%) (Alvisi *et al.* 1996). Above this was a layer of black fine grained gyttja. Other cores had a similar structure, with the thickness of the upper layer varying from 10-15 cm depending on core location. A number of lighter and darker bands are present throughout (Frignani *et al.* 1995), though these are not annual laminations. As in most sub-alpine lakes (e.g., Maggiore, Mergozzo; Guilizzoni & Lami 1992) with large drainage basins, the pelagic lacustrine sedimentation at the maximum depth is often interrupted by turbidites (Guilizzoni & Lami 1990). Between 5 and 8cm there are a number of turbidites that have been related to historical flood events in 1963 and 1965 (Ambrosetti *et al.* 1994). In the shallower Buccione Basin (water depth 30 m) the sediments are more homogeneous, although with lower sedimentation rates than in the deeper Pettenasco

Basin (cf. Alvisi 1993; Tab. 3). Loss on ignition is rather high ranging from ca 10% d.w. to 40% during 1960-1990. Carbonate concentrations are low (ca 7%).

5. DATA ANALYSIS

The sediment sequence in core OR1 was split into 5 broadly matching zones based on the magnetic susceptibility (Alvisi *et al.* 1996) and pollen results (see below; Alvisi 1993). Zone I extended from 88-c.67 cm, zone II from c.67-45 cm, zone III from 45-ca20 cm, zone IV from 20-10 cm, and zone V from 10-0 cm.

Core OR-92 was zoned using CONISS (CONstrained Incremental Sum of Squares) cluster analysis of the geochemical and pigments profiles with square root transformation of the original data to optimise the signal to noise ratio. In this scheme zone I extends from 70-20 cm, zone II from 20-9 cm, zone III from 9-4.5 cm, zone IV from 4.5-1.75, and zone V from 1.75-0 cm. The sequence of pollen grains was divided on a preliminary and subjective basis into five broad biostratigraphic zones which match reasonable well to the CONISS plot for the sequence.

Comparing the different phases of the lake development based on this analysis (cf. Figs 5 and 6 below) it appears that the OR-92 zone I corresponds to OR1

zones II and III; OR-92 zone II corresponds to OR1 zone IV; OR-92 zones III, IV and V correspond to OR1 zone V.

6. MINERALOGY

Mineralogical analysis of the core sediments can be used to determine the nature and amount of the sedimentary input eroded from the drainage basin as opposed to that derived from physical-chemical processes within the lake. Bulk dry sediment and the clay fraction from core OR1 was analysed by means of an X-ray diffractometer (see Alvisi 1993 for methodological details). Due to analytical problems, in zone I (88-67 cm) only the clay fraction was analysed. In figure 5, the mineralogical data for this zone refer only to that fraction whereas for the other zones the data refer to the bulk sediment.

The results are based on the heights of selected peaks of reflection of the main mineral phases (Tucker 1988). Derived parameters that are more sensitive to environmental changes (e.g., the illite crystallinity index; Chamley 1989) are also discussed below. The inorganic fraction, calculated as the residue in percentage of the dry weight after loss on ignition at 550 °C (ash content), is about 88% d.w. in the lower zones, I to IV, with only two exceptions at around 67 (85-86% d.w.) and 45 (91-92% d.w.) cm, and decreases to 75% in the uppermost zone V.

The mineralogical composition of the Lake Orta shows two main phases of lake development (Fig. 5): the first phase, from the core bottom to 10 cm depth (spanning zones I-IV), reflects the meso-oligotrophic condition typical of other Italian sub-alpine deep lakes (Guilizzoni *et al.* 1983); the second, from 10 cm to the top of the core (zone V), is marked by sharp variations in many parameters clearly related to the strong chemical modifications in both the water column and sediments compartments that occurred during the main period of pollution.

Each of the zones within these two main phases is characterised by a number of mineralogical features based on changes in the quality and amount of sedimentary input to the lake bottom.

Zone I (88-67 cm). Here it is possible to observe few changes in the composition of the sedimentary input to the lake with a substantial stability of the accumulation rates as indicated by the susceptibility curve. The clay fraction shows the presence of kaolinite, the absence of degraded illite even with a high mica content, and a slight upward increase of detritic phases such as quartz and plagioclases. The crystallinity index of the total illite does not show any changes. However, the index of micas and degraded illite taken separately show a different behaviour. At the bottom of the profile the micas seem to be not degraded, but then the crystallinity of the micas decreases and the degraded illite begins to accumulate, probably favoured by an increase of the hydro-

logical activity in the drainage area and increased erosion. The further increase of the illite degradation and the contemporary increase of the crystallinity index suggest an input of both weathered and unweathered material. All these features suggest a possible climatic amelioration from cooler and wetter to increasing warmer conditions from the bottom to around 75 cm depth of the core and/or an opening of the canopy with a consequent exposition of soils and rocks to weathering. In fact, the first increase of degraded illite input followed by the increase of primary micas could be due to the deeper erosion of the exposed areas with a consequent transport to the lake of both mineralogical components without any further chemical weathering. In the second part of this zone the detritic phases seem to decrease while the degraded illite still increase at the expenses of micas which disappear, and the organic fraction increase slightly around 67 cm. These features suggest a decrease of deep erosion and a shift toward more organic sediments probably due to a recovery of the vegetation over the exposed areas possibly favoured by increased wetter conditions as shown by the pollen assemblage.

Zone II (67-45 cm). The dominance of detritic phases such as quartz, micas, augite and degraded illite (and the increase of the magnetic susceptibility values), average values of which are higher than in the other zones, points to an increase in the inorganic fraction sedimentation rate. Two major phases of deep erosion of unweathered material from the slopes are evident: the first one around 65 cm and then, after a period of relatively constant erosion rate, a second starting from 52 to 45 cm. The pollen diagram shows that these increases are mainly the result of disturbances on the lake shores and slopes, linked to agricultural purposes. The land management changes seem to take place inside the drainage basin of the lake, its effects being recorded very rapidly in the lake sedimentary column.

Zone III (45-20 cm). The rather constant concentrations of organic matter, the decrease of the concentrations of the major indicator elements of erosion, and the uniform mineralogical composition point out a period of decreasing sedimentation rates. The ratio between organic and inorganic components remain unchanged during this interval. The susceptibility curve shows an initial decrease up to 35 cm, followed by quite constant values similar to those preceding the peak at 45 cm depth. However, a slight increase of phosphorus, sulphur and silica, showing a different behaviour with respect to the detrital elements, should be linked to the increase of their organic compounds and thus could represent an increased productivity of the lake. All these features suggest a partial recover of the vegetation leading to a new equilibrium between erosion and vegetation cover, but the mean sedimentation rates are still higher than those in with Zone I.

Zone IV (20-10 cm). At the beginning of this short interval the mineralogical composition shows a sharp increase of detrital phases such as quartz, plagioclases and micas, followed by an important drop in the detrital fraction and again a new recovery upward. This feature suggests a short period of rapid input of sediment to the lake probably due to a localised disturbance on the lake shore such as excavation for building purposes.

Zone V (10-0 cm). The huge peak of the susceptibility curve and the general increase of almost all the geochemical and some mineralogical parameters at the beginning of this interval point out a strong modification of the lake environment probably leading to particular stressed physical-chemical conditions on the lake floor. Among the detrital phases only quartz show a clear increase, whereas the others seem to decrease slightly probably because of their instability, except quartz, in the new lake conditions. The clay fraction also increased together with pyrite and sulphur (gypsum) revealing a more reactive mineralogical environment with possible oxygen depletion on the bottom. The analysis of the surface sediments reveals very high concentrations of copper, chromium, sulphur and iron. These elements were probably precipitated into the sediments soon after the pH of the lake waters was increased by the recent liming treatment directed to improve the quality of the lake basin (AA.VV. 1990, Calderoni *et al.* 1991). All the other elements display higher concentrations and for some of them the causes are the same as those seen above. Also the increased concentrations of some elements such as Mn, Ca, Mg, Al, Fe and Ni at the core top (see below) could be the result of the composition of the calcium carbonate used for liming operations.

7. GEOCHEMISTRY

Major elements (Ca, Mg, Na, K, Si, Al, Fe, Mn, S) were sparsely measured on core OR1 (collected in 1990 in the Southern Basin, Tab. 1) (Alvisi 1993). Here we report on results from that and other studies (see below).

Major cations. Element such as calcium, magnesium sodium and potassium in lake sediments are primarily associated with allogenic clastic eroded material from catchment soils and rocks. In particular, K (1.6-2.6% d.w.), Na (0.8-1.3% d.w.) and Mg (1.6-2.7%) are often used to detect past soil erosion rates (Engstrom & Wright 1984). Period of higher erosion are shown from the profiles of these elements (e.g. 1963, 1965, Tab. 1; cf. Guilizzoni & Lami 1990). Si (54-62% d.w.) and Al (12-21% d.w.) trends show opposite behaviour, whereas Ca does not fluctuate through time (*ca* 0.7% d.w.) except an increase during the early 1990s (1.3% d.w.) (Alvisi 1993).

Fe, Mn. These two elements are perhaps the most important for palaeolimnology although their profiles are often difficult to interpret. In fact, a number of inde-

pendent environmental factors control the supply of Fe and Mn to the lake sediments (e.g. erosion, weathering from the rocks, soil composition). Mackereth (1966) was probably the first to clearly recognise that their different mobility can be used to infer palaeo-redox conditions.

In Lake Orta the profile of Fe concentration shows a regular decreasing trend from 8% d.w. at the beginning of 1900 to *ca* 5% at the end of the 80s. Mn decreases instead sharply from *ca* 1950 (from 1000 ppm to 250 ppm) (Guilizzoni & Lami 1990; Muntau, unpublished data). These metals undergo resuspension and migration when sediment-water interface becomes reduced through oxygen depletion. The Fe/Mn ratio increase steadily from 1960 (in coincidence with the increase oxygen hypolimnetic deficit) to the late 1980s (Guilizzoni & Lami 1990).

Sulphur. Before 1920-30 this element has a mean concentration of *ca* 0.20% d.w. Then it increases to a maximum (3% d.w.) in the early 1990s. There is a general agreement between this profile, the carbonaceous particles (Fig. 6) and magnetic curves (Alvisi 1993; Frignani *et al.* 1995) that may reflect a major increase in the atmospheric contamination by fuel combustion. In addition, there are many evidences from the Fe and Mn levels and the evolution of trophic and oxygen concentration (Calderoni & Tartari 2001), that the enhanced S accumulation in the topmost *ca* 15-10 cm of the core could be also related to the precipitation of ferrous sulphide which gives to the mud the characteristic black colour.

Nutrients. The concentrations of organic carbon and organic nitrogen exhibit a marked, progressive increase upward across the 1920 horizon (C: from *ca* 4% d.w. to 20%; N: from 0.4% to 2.4% d.w.; Guilizzoni & Lami 1990). These elements are accumulated in high quantities between 1930 and 1950: accumulation rates and concentrations increased from 70 (4% d.w.) and 10 g m² y⁻¹ (0.4% d.w.) to 170 (10% d.w.) and 20 g m² y⁻¹ (1.4% d.w.), respectively (Guilizzoni & Lami 1990).

The C:N ratio indicates a prevailing lacustrine contribution of organic matter (values around or below 10; Guilizzoni & Lami 1990).

Also total phosphorus exhibits a similar marked increase during the last 50 years ranging from a baseline content of *ca* 0.22% d.w. to *ca* 1% d.w. Taking into account all the pollution history in Lake Orta, variation in P retention which is controlled by factors such as redox potential, Fe chemistry, pH (Mitchell *et al.* 1990) – is in this lake probably more important than changes in P loading.

Other metals. Since 1958 many analyses of metals (particularly Cu) were performed in sediment cores of Lake Orta (e.g., Provini & Gaggino 1985; Guilizzoni & Lami 1990; Alvisi 1993; Muntau *et al.* unpublished data) and the paper by Baudo & Beltrami (2001) will give an ac-

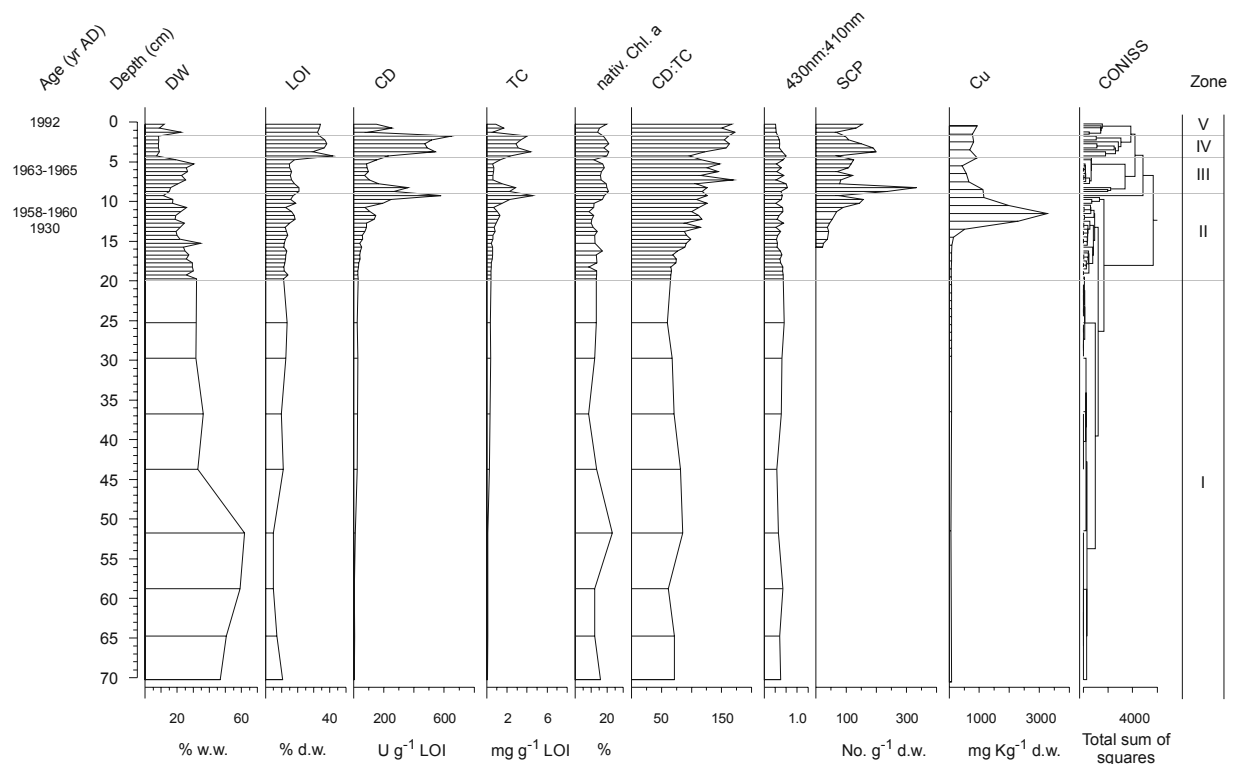


Fig. 6. Lake Orta, core OR-92. Selected physical and geochemical parameters. DW = dry mass; LOI = Loss-on-ignition; CD = Chlorophyll derivatives (U: absorbance units); TC = total carotenoids; Nativ. Chl-*a* and 430nm:410nm ratio = pigment degradation index; SCP = carbonaceous particles; Cu = copper. CONISS zones I-V are shown.

count of these studies. Here we report the Cu profile from core OR-92 (Fig. 6). All analyses clearly showed the many fold increase of the "baseline" heavy metal concentrations, (e.g., for Cu and Cr the natural concentrations are set at *ca* 80, 40 ppm dry weight, respectively) a few years after the beginning of the rayon factory's discharge (Guilizzoni & Lami 1990). In particular, the Cu sediment profiles parallel that on lake water at winter turnover (Cattaneo *et al.* 1998), showing a sharp decrease after 1958 (from *ca* 3300-5000 ppm d.w. to 1000 ppm d.w.; Fig. 6) when the discharge was drastically reduced. Similarly the Cr concentration shows a sharp increase from a baseline value of *ca* 40 ppm to a maximum at the end of the 1950s of *ca* 4000 ppm. Because of the acidic bottom water conditions the concentrations of Cu, Cr and Al in the water column reached very high values (around $100 \mu\text{g l}^{-1}$) during the early 60' and between 30 and $50 \mu\text{g l}^{-1}$ from mid 1970 to 1985 (Calderoni & Mosello 1990; Calderoni & Tartari 2001).

8. ORGANIC MATTER AND FOSSIL PIGMENTS

Water content is commonly highest at the top of the core (*ca* 80%). Fluctuations are the result of turbidities as is the case for the sharp decrease in OR-92 at 5-8 cm (Tab. 2-3; floods events in 1963, 1965; Ambrosetti *et al.* 1994) which dilute all the organic compounds (see below).

Loss on ignition increases but with some fluctuations, from less than 6-10% d.w. in the oldest sediments to *ca* 40% in the period 1965-1992 (core OR-92; Fig. 6), contemporarily to the establishment of eutrophication. However, as for the biological records, the significant increase of LOI begins around 1920-30 (values of *ca* 20% d.w.), when senescent or dead organisms began to settle and accumulate because of pollution (Monti 1930).

An index of pigment preservation is given by the native chlorophyll, i.e. the proportion of chlorophyll not degraded to phaeopigments (Swain 1985). This index increases following the onset of lake pollution from a pre-1930 average of *ca* 15% to 19-20% (Fig. 6). We postulate it is related to lake acidification and oxygen consumption through NH_4 oxidation (Adams *et al.* 1978; Guilizzoni & Lami 1990), which conditions inhibited microbial break-down of pigments (Guilizzoni & Lami 1988).

In any pigment study the question "are the changes related to a better preservation or/and to the variations in pigment production?" is of central importance (Leavitt 1993). In Lake Orta these two factors seems to operate jointly. First, the increases of chlorophyll preservation, as shown also by the 430nm:410nm ratio (Guilizzoni *et al.* 1993), do not always match the rise in pigment (Fig. 6). Second, there is an earlier period of

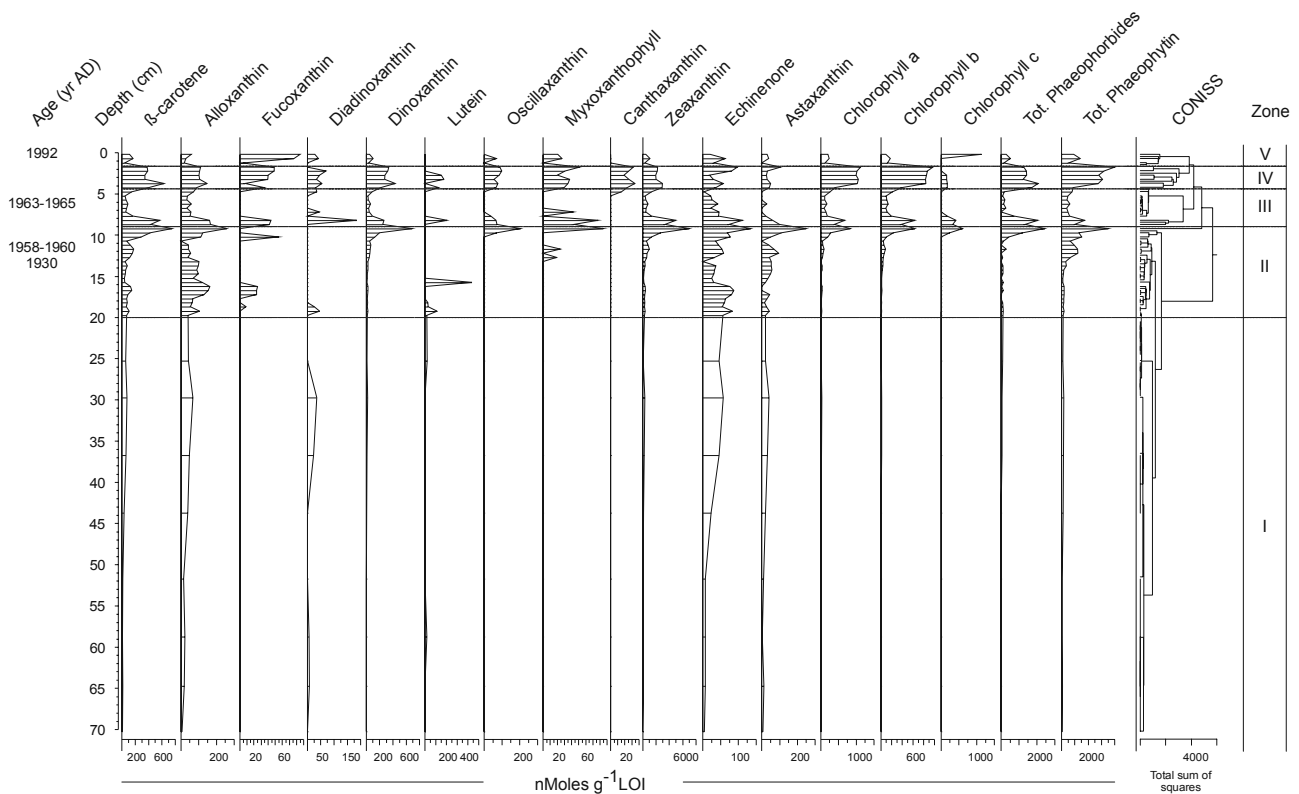


Fig. 7. Lake Orta, core OR-92. Algal fossil pigment concentration profiles. CONISS zones I-V are shown.

good preservation (*ca* 52 cm) during which pigment concentrations do not change.

Sedimentary CD and TC are detected in large amounts from 1930 onwards (Fig. 6). Their concentrations in the last *ca* 60 years are of the same magnitude as those detected in eutrophic lakes. Because of the toxic effect of Cu the post-1930 rise must have been caused mainly by a massive mortality of the phytoplankton community. The sharp decrease in all the pigment concentrations between 5 and 8 cm is due to large quantities of mineral deposition from soil erosion (floods of 1963 and 1965) that diluted the organic content of the samples. Notably low are total pigment concentrations in the pre-1900 periods. Moreover, the similarity in CD and TC curves indicates little allochthonous pigment input.

Analysis of single fossil carotenoids suggests that there have been dramatic changes in algal abundance and composition during the pollution decades (Zones IV and VI; Fig. 7). Concentrations of the chemically stable carotenoid β -carotene, indicative of total algal biomass, is variable in this period and shows little trend with depth: a slight rise in concentration is observed at *ca* 30 cm. Here also diadinoxanthin (siliceous algae), alloxanthin (cryptophytes), echinenone (cyanobacteria) and astaxanthin/astacene show a peak. As a whole, the pre-1900 concentrations (zones I and II) are extremely low indicating a low productive environment. However, the increase in-lake productivity can be dated before the

pollution onset (1930; zone III). The effect of early century industrialisation human disturbance in the Lake Orta catchment area is accordingly seen also from the carbonaceous particles (Fig. 6), Pb and S profiles (Alvisi 1993), as well as from pollen analysis.

Cyanobacteria are always present: they are particularly abundant from 50 cm upward. Colonial species are present during recent times only (zones III-VI), where very abundant is also zeaxanthin (carotenoid characteristic of green algae and cyanobacteria). Peaks of carotenoids are shown in zone IV and VI. These very variable pigment trends also support the evidence of an extremely biologically unstable ecosystem during the pollution phase.

9. POLLEN

For pollen analysis 11 sediment slices 1.5 cm thick were sampled every 8 cm in core OR1. After adding a known amount of *Eucalyptus* pollen grains, the samples were prepared following the procedure described by Faegri & Iversen (1964) and modified by Alvisi (1993). A minimum of 500 terrestrial pollen grains have been counted for each sample. The grains have been identified on the basis of key tables (Faegri & Iversen 1964; Moore & Webb 1978), and reference material. The data have been elaborated and plotted with TILIA software package (Grimm 1991) with five categories of terrestrial and aquatic taxa: trees, shrubs, herbs, aquatic and spores.

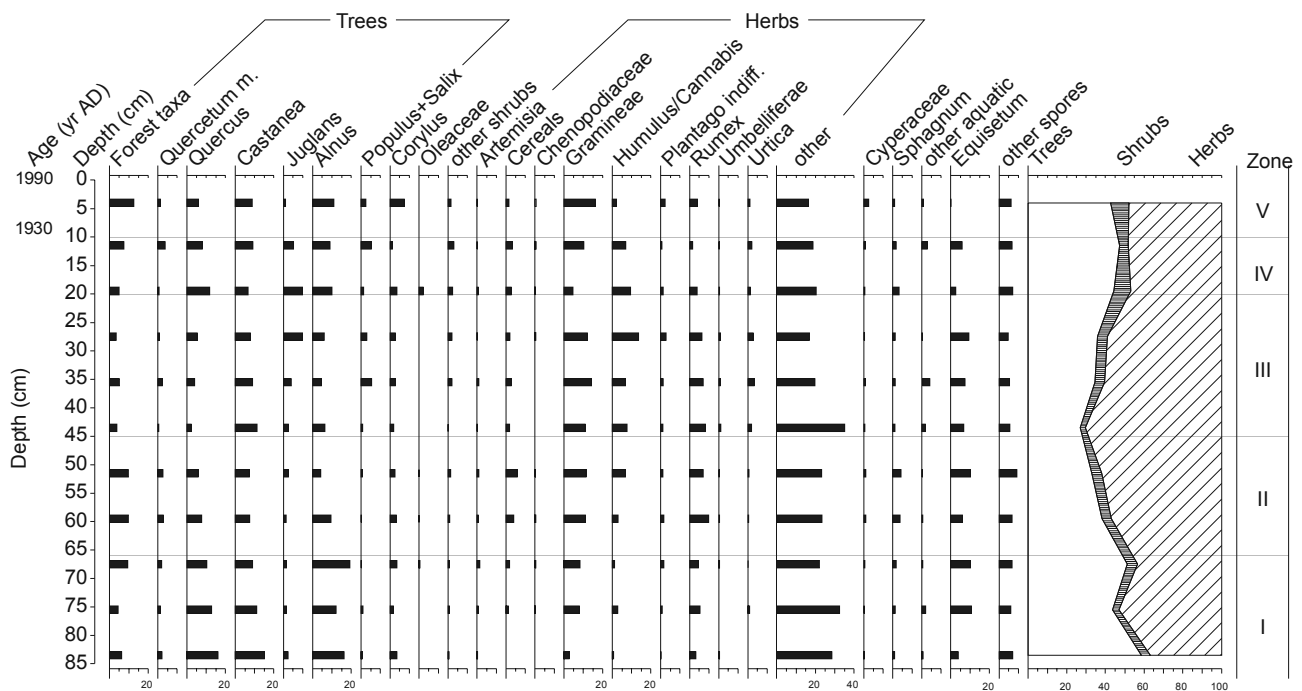


Fig. 8. Percentage pollen diagram (principal taxa only) for the core sequence OR1 from Lake Orta.

Pollen sequence can be broadly subdivided into five biostratigraphic zones (Fig. 8) closely corresponding to those defined by the mineralogy and magnetic susceptibility records.

Zone I (88-67 cm). This zone shows a slight decreasing trend of the arboreal vegetation without any evidence of agricultural indicators (cereals and ruderals) from the core bottom to around 75 cm. The opening of the canopy should then be natural (dryness, fire) or possibly due to wood exploitation by local population. Therefore, we would expect an increase of the runoff and thus erosion, but the pollen diagram shows that the major decrease is at the expenses of forest taxa and less of the oak wood. This means that the decrease in forest cover should have taken place at middle to high altitudes and the oak forest was still present at low altitude preventing an increase of sediment input to the lake. Above 75 cm depth, the arboreal percentages, and in particular the forest taxa, increase toward previous values and even higher. The taxa linked to humid environment increase as well suggesting an increase of the precipitation on the area with a possible drowning of some low land and formation of small peat bogs and swampy areas. The drop of some major and minor elements concentrations suggests that the increase of the forest canopy and the presence of aquatic taxa prevented an important sediment contribution to the lake.

Zone II (67-45 cm). The pollen diagram shows a clear progressive fall of the arboreal taxa mainly at the expenses of *Quercus* and *Alnus*. The forest taxa (*Betula*, *Fagus* and *Pinus*) are present up to 50 cm depth sug-

gesting cool and wet climatic conditions. The most interesting feature of this zone is the presence of cereals, showing the highest value (max at ca 50 cm). Together with other ruderals, which are indicators of human activity (*Compositae*, *Plantago*, *Rumex*), these features point out the beginning of important agricultural activities in the catchment or an increased effect of the agriculture in the nearby Po Plain probably favoured by suitable climatic conditions. Although the progressive fall of taxa linked to wet environment (*Equisetum* and *Sphagnum*) suggests a decrease of precipitation in the area or a destruction of the vegetation along the lake shores, the slightly fall of oak wood suggests a widespread clearance of the drainage area as the main responsible of the crop abandon. The deforestation, however, should have been still the consequence of a new climatic change favouring now a different agricultural activity. In the pollen diagram, in fact, it is possible to observe the beginning of the rise of *Juglans* frequencies from the end of this zone. Nowadays, all the slopes around the lake are still covered by many walnut and sweet chestnut trees.

Zone III (45-21 cm). The pollen assemblage reflects the presence of a woody cover characterised by an open canopy with an important presence of Gramineae and *Humulus*, testifying a remaining human influence on the plant association following the abandonment of cereal crops in favour of arboreal plantations. Poplar, willow, *Urtica* and part of the aquatics show a slight increase only around 35 cm suggesting the fast recovery of these wet taxa on the lake shore in advance of the slower woody taxa. Some trees (*Quercus*, *Alnus*, *Fagus*) and

shrubs (*Juniperus*, Oleaceae) show an increase in the uppermost part of this interval whereas hop and weeds, *Equisetum*, *Plantago*, *Rumex* show a decrease during the same period. These changes suggest a further recovery of woody taxa at the expenses of pioneer and ruderal species.

Zone IV (21-10cm). The pollen assemblage reflects a new perturbation of the forest ecosystem: *Quercus*, *Alnus* and *Juglans*, Oleaceae and *Corylus* all decrease together with *Rumex* and *Humulus*. At the same time, the recovery or the increase of many different taxa, among them herbaceous taxa and less-demanding trees, suggest an artificial diversification of the vegetation due to a new clearance of some areas with a consequent opening of the canopy. The modification of the plant cover seems to be mainly directed toward lowland and coastal areas probably caused by an increase of population around the lake shore.

Zone V (10-0cm). The high diversity of the vegetational assemblage coupled with low single frequencies suggest the presence of continuous disturbances on the vegetation, probably enhanced by the constant increasing exploitation of the lake shores and slopes by people. The disturbance seems to determine a further increase of erosion and thus of sedimentation rates as also suggested by the increase, at the beginning of this interval, of the concentrations of quartz, plagioclases, k-feldspar and micas and detrital phases such as K, Na and Mg.

10. EFFECT OF POLLUTANTS ON BIOTA

10.1. Changes in size and species composition

Manca & Comoli (1995) reported results on fossil Cladocera assemblages in a 37 cm long sediment core collected in 1994 (OR-94) at a station located in the south basin of the lake (Buccione; Fig. 1). Because of the low sedimentation rate almost 400 years were recorded in this core, though this still allows the possibility of a fine resolution (1 cm sediment integrates 5-10 years during the most remote period).

The availability of over 100 years of literature information also allowed the comparison of results on fossil Cladocera with studies on lake fauna and large-scale perturbations. They reveal three phases in the lake's history: an earliest one where the original community was present (17th Century), an intermediate one reflecting eutrophication and acidification, and the most recent one in which the initial recovery was detectable.

The zooplanktonic community before the 19th century indicates a lake with low productivity and low alkalinity. Chydorid species diversity was about 2 (1.86 ± 0.21) and evenness was close to 1 (0.91 ± 0.05). Planktonic Cladocera were most represented by *Eubosmina longispina* and *Bosmina longirostris*, *Daphnia longispina* group, and especially *Sida crystallina*. At that time, in the deep sub-alpine lakes of Italy and Switzerland, *S. crystallina* developed truly pelagic populations, of a morphotype having the adhesive gland atrophic

(var. *limnetica*; Hutchinson 1967; Margaritora 1983), and able to live down to 150 m depth (Baldi *et al.* 1953).

A series of events affected the Cladocera community. Apart from the earliest changes (probably driven by changes in temperature, see below), the first event to modify the original community of the lake is detected during the period in which exotic fish (landlocked shad, *Alosa phallax nilotica*) were introduced into the lake, accomplished in 1880 (De Agostini 1897). The total number of remains increased, and of the two subgenera of *Bosmina* present before, only *Eubosmina* was left for a while. This has been interpreted as a result of the positive selection of *Bosmina* over *Eubosmina* by the *Alosa phallax lacustri* (or *nilotica* [?]), because of its habit of preying in the upper 20 m, as observed in Lake Maggiore (Berg & Grimaldi 1966). *Eubosmina* is in fact usually found in deeper waters (Margaritora 1983).

There was a good correspondence between the planktonic Cladocera community described in 1879 by Pavesi and the assemblage reconstructed from the remains found in the core sections at this level. Both the *longispina* and the *pulex* groups were present within the nominate subgenus of the *Daphnia* genus. Although only *Bosmina longirostris* was reported by Pavesi (1896), in the sediments *Eubosmina* was in general more abundant than *Bosmina*.

The phase of severe pollution (1928-1960) is first detected in the core as a dramatic decrease in the Shannon diversity index and in the number of species of Cladocera: *Chydorus sphaericus*, initially with some *Alona* are the only taxa left at this level of the core. The decrease in the pelagic/littoral ratio, down to 20%, indicate that planktonic species were more affected than the littoral ones. Total concentration of remains also decreased dramatically with pollution (Manca & Comoli 1995). The drastic changes in the zooplankton community was also followed, with a delay of some years, by the disappearance from the sediments of cocoons belonging to the oligochaete *Spirosperma ferox* (Bonacina *et al.* 1986).

The earliest sign of improved conditions in Lake Orta may be the increased accumulation of organic matter in sediments between 1951 and 1962. An increase in Cladocera remains has been detected only from 1962, after the initial recolonisation by phytoplankton. Vollenweider (1963) described the lake in the period 1958-1961 as highly unstable, with rapid recolonisation by various phytoplankton species, which attained high values of total biomass. No Cladocera were found in the pelagic zone. Due to the tremendous increase in ammonium concentration and the high estimates of primary production, he argued that the lake was undergoing a process of acidification and eutrophication. Indeed, fossil pigments of this period show variable but generally high accumulation rates with the presence of *Oscillatoria* sp. (Figs 6, 7).

The sediment core indicates that *Chydorus sphaericus* is the dominant species during the recolonisation phase. Although being typically littoral, at the beginning of the 1980s it developed a truly pelagic population in the lake (Bonacina & Bonomi 1984), thus suggesting the presence of abundant filamentous algae and blooms of blue-greens in the pelagic zone (Hutchinson 1967). This species is known for its ability to become planktonic, in lakes rich in filamentous algae, which are used as a feeding substrate by the animals (Frey 1968).

The recolonisation of the lake by truly pelagic species is also detected in the core, with a rise of the P/L ratio above 50%, and a decrease in chydorids. The general increase in the number of remains of the uppermost 6 cm of the core also reflects an increase in their accumulation rate.

10.2. Effects of metal contamination on diatoms

Lake Orta has also provided an opportunity to study impact of metals pollution on diatom assemblages. Ruggiu *et al.* (1998), in a palaeolimnological study described the acute and long-term effects of Cu contamination on the diatom community of the lake, at a water concentration of about $80 \mu\text{g l}^{-1}$. They documented the progressive impoverishment of the diatom community of the lake, which was originally similar to that of the other deep, subalpine lakes. *Fragilaria crotonensis*, *Asterionella formosa*, *Cyclotella bodanica* and *C. stelligera* were quickly extirpated by pollution and never reappeared. Species like *Synedra tenera* were more resistant to pollution, and reacted with an increase in the teratological forms, already present in the lake, at very low percentages. This observation confirmed the link between diatom teratogeny and metal pollution (Yang & Duthie 1993; McFarland *et al.* 1997). During pollution, *Achnanthes minutissima*, "the most important diatom species both in the sediment and the plankton of the lake" (van Dam & Mertens 1990) is positively selected, confirming its tolerance to high levels of metals, copper included (Takamura *et al.* 1989; van Dam & Mertens 1990).

10.3. Changes in the size structure of communities at different trophic and phyletic levels

Cattaneo *et al.* (1998) applied a size-based approach (Peters 1993) to the analysis of the fossil remains of Lake Orta in the same core (OR-94) used by Manca & Comoli (1995), following the size distribution of diatoms, thecamoebians, and cladocerans of about 300 years before the onset of pollution to 60 years afterwards. They interpreted Lake Orta pollution as a stress in the sense of Odum (1985), who theorized the dominance of small, rapidly reproducing organisms, in stressed ecosystems. The reduction of average size in the three communities at different phyletic and trophic levels, as well as the selection of smaller morphotypes within a taxon, is interpreted as a validation of the hy-

pothesized link between stress and size. Since planktivorous and piscivorous fish were both practically absent, the success of smaller organisms following the onset of pollution could not be explained by selective predation, but was likely to depend on the increased resistance of smaller organisms to contaminants. The toxic doses of many contaminants decline with size (Chappell 1992), and perhaps the longer generation time and lower clearance rates of larger organisms (Fenchel 1974) expose them to effectively higher levels of pollutants, resulting in greater mortality. Previously, the link between size and stress had been demonstrated only in short term, laboratory tests. Lake Orta has provided a unique case to study the response to a chronic pollution that lasted over 50 years.

11. EFFECT OF CLIMATE CHANGE

A study of the remains of different type of organisms in the 37 cm long core OR 94 revealed that an important environmental change occurred in the second half of the 17th century. Changes in size and species composition of thecamoebians were indicative of a decrease in water temperature (Asioli & Medioli 1994). The concomitant decrease in concentration and accumulation rate of Cladocera, with a shift toward littoral species (decrease in Planktonic/Littoral species) was also regarded as indicative of a decrease in water level driven by a decrease in temperature (Manca & Comoli 1995). Changes in habitat diversity at stable water levels can conceivably bring about the same changes (Frey 1986) and changes in the P/L ratio can result from a differential response of the planktonic and littoral Cladocera to changing primary production, with planktonic forms favoured when productivity increases (Crisman & Whitehead 1978). Indeed, decreases in total abundance and accumulation rate of remains are generally interpreted as indicating lower temperatures, since animals moult and reproduce less when cold (Frey 1986). However, a 1 m core taken from the deepest area showed that during the 17th Century there was a significant decrease of SiO₂, S, P, and generally lower concentrations of algal pigments (Figs 6, 7) compared with those recorded for the 18th and 19th centuries.

Palaeoclimatic studies do show a general decline in temperature of 1° C world wide during the time of these changes in the community structure in Lake Orta (1650-1675), a period known as "Maunder's minimum" (Lamb 1977; Eddy 1976).

12. CONCLUSIONS AND FUTURE RESEARCH NEEDS

It has been shown by the above chapters that a number of palaeolimnological studies were carried out during the past 10-15 years and how the results served to integrate the existing data and to add new information on the long-term limnological record of this lake case study. The gradual enormous loading of copper and

ammonium salts has had a profound effect on the limnological history of Lake Orta. The chief consequence of the pollution was a nitrate and ammonia accumulation, a striking lowering of the pH and an increased oxygen hypolimnetic deficit. The manifestation of this in the sediments has been initially a general marked decrease or even a complete disappearance of the invertebrate organisms and diatoms, an increase of pigments (and LOI) due to senescent or dead algae accumulation, and subsequently a strong modification in their taxonomic composition and assemblages. However, the process has been neither regular nor progressive: there have evidently been several large fluctuations in productivity and organism biomass. Deep changes in the reduction of average body size in three communities (diatoms, thecamoebians, and cladocerans), and the presence of teratological frustules of *Synedra tenera* during the 50 years of pollution, were also reported. In 1989-1990 the lake was treated with lime and it rapidly recovered. Unfortunately, even the most recent studies (Manca & Comoli 1995; Cattaneo *et al.* 1998; Ruggiu *et al.* 1998), were made on cores collected in 1994 and the core sectioning adopted (usually 1 cm, corresponding to 3-5 or more years; cf. Tab. 3) had not a sufficient resolution for studying in detail the sub-fossil remains in relation to the lake recovery by liming. Future high resolution sediment core studies (possibly at annual resolution) should be addressed in the direction of consider this large impact event not only or as much to monitor the biota development and recovery as to compare and "calibrate" these sediment data with the well documented last 12 years of limnological information.

As regards the "natural variability", climatic changes during recent centuries (e.g. the Little Ice Age) only agree in a very general way with primary production rates. Changes in Cladocera density, size and composition, and pigments concentrations appear to occur before climatic or anthropogenic changes are indicated by the pollen record. Thus, changes in the pollen composition appear to be a very conservative indicator of climate change. Also in this case, however, without a finer resolution study around the main biological and geochemical changes in the cores it is difficult to determine the exact sequence of events associate with natural impacts.

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